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## Seasonal changes of sources, processes, and volatility of organic aerosol at urban, coastal and forest sites in Eastern Europe

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The stable carbon isotope  $^{13}\text{C}$  has the potential to give insights into sources and processing of organic aerosol. However, the use for source apportionment has been somewhat limited, because the  $^{13}\text{C}$  source signatures vary and show some overlap.  $^{13}\text{C}/^{12}\text{C}$  ratios are usually reported as  $\delta^{13}\text{C}$  indicating a permil deviation from the international reference standard Vienna Pee Dee Belemnite (V-PDB).

We use a method to measure  $\delta^{13}\text{C}$  in OC desorbed from filter samples at three different temperature steps: 200 °C, 350°C and 650°C (Zenker et al.,2020). The results give a rough indication of aerosol volatility, as more volatile compounds usually desorb at lower temperatures.

We demonstrate with an extensive source study that in Lithuania and likely other Eastern European regions, the main anthropogenic primary sources for organic carbon (OC) have distinct isotopic signatures.  $\delta^{13}\text{C}$  values of vehicular emissions show the most negative values around - 29 ‰, emissions from combustion of the most common wood types are more enriched with values around -26 to -27 ‰, and coal burning is around -25‰. For source samples  $\delta^{13}\text{C}$  values at the three desorption temperature steps usually do not differ more than 1 ‰.

For ambient aerosol samples, the differences in  $\delta^{13}\text{C}$  values at different desorption temperatures are usually larger. This indicates varying source contribution or different chemical processes leading to the different volatility fractions. Combined isotopic and chemical analysis showed that in winter was a clear distinction in source contribution between the less refractory OC and the more refractory OC. We were able to identify fossil fuel burning as predominant source of the less refractory OC in the small particle size range ( $D < 0.18 \mu\text{m}$ ), and biomass burning as predominant source of the more refractory OC in the larger size range ( $0.32 < D < 1 \mu\text{m}$ ).

At all three sites, OC had more negative  $\delta^{13}\text{C}$  values in summer than in winter which can be explained by the contribution of biomass/coal burning sources in winter. At the urban site  $\delta^{13}\text{C}$  of OC did not change much with increasing desorption temperature in winter, which is typical for primary sources. In the summer  $\delta^{13}\text{C}$  of OC was clearly more negative for lower desorption

temperatures at all three sites. This is likely due to the influence of secondary organic aerosol formation in summer, which should have depleted (more negative) isotopic signature and contributes strongly to the more volatile fraction.

A higher fraction of more refractory OC in summer compared to winter-time suggests active photochemical processing of the primary organic aerosol as an important process at all three sites. During a pollution episode transporting aged pollution from Poland and southern Europe to the otherwise clean forest site, a potential isotopic signature for photochemical aging was identified.